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ULTRASONIC ATTENUATION IN MnF_2 NEAR THE NÉEL TEMPERATURE*

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Changes in many physical properties including internal friction and Young's modulus have been measured near the Néel temperature for several antiferromagnetic materials. The mechanical properties of the metal chromium have been extensively investigated¹⁻⁵ and show anomalies at the Néel temperature (310°K) and at "spin-flip" temperature (120°K). Recent measurements^{4,5} show a dip in Young's modulus at the Néel temperature and a change in internal friction which starts at the Néel temperature and becomes a maximum at the "spin-flip" temperature. These results, along with the relatively small heat capacity anomaly² in chromium, are in agreement with Overhauser's treatment⁶ of the antiferromagnetism of chromium as spin-density waves rather than as localized moments.

Experiments on oxides of cobalt,^{7,8} nickel,⁸ and manganese⁹ show similar effects: a lowering of modulus and an increase in internal friction as the temperature is lowered past the Néel temperature. For MnO and CoO , changes in modulus and well-developed peaks in internal friction are observed and found to occur at temperatures somewhat below the Néel temperatures. A thermodynamic treatment⁹ predicts that the internal friction peak and the change in modulus always occur together and that the maximum in the internal friction occurs at a temperature lower than the Néel temperature.

In this Letter we present preliminary results of ultrasonic attenuation studies on single-crystal MnF_2 which differ from those cited above and are characterized by the following: a frequency-dependent attenuation peak for longitudinal waves only, which occurs very close to the Néel temperature; relatively slight dependence of attenuation on magnetic field; and no observable change in elastic constant at the Néel temperature. Measurements have been made of the attenuation and velocity of ultrasonic waves propagating parallel to [110] for temperatures between 58 and 90°K.

Both longitudinal and transverse waves were studied at frequencies up to 65 Mc/sec. The room-temperature path length was 1.386 cm.

Short rf pulses were supplied by an Arenberg pulsed oscillator which was connected to the sample, receiver, and time-mark generator in the usual manner.¹⁰ Absolute values of wave velocity were determined from the room-temperature length as corrected for thermal expansion¹¹ and the measured time between successive pulses. Although it was not possible to determine a complete set of moduli, the values at 70°K of the constants ($\frac{1}{2}C_{11} + \frac{1}{4}C_{12} + C_{66}$), C_{44} , and ($\frac{1}{2}C_{11} - \frac{1}{2}C_{12}$) are, in units of 10^{11} dyne cm^{-2} , 16.92, 3.257, and 1.019, respectively.

Changes in wave velocities were determined by observing the change in transit time of an echo which had undergone multiple reflections. Changes in transit time of 0.01 sec could be detected out of a total transit time of over 50 sec, thus making a relative accuracy of determination of wave-velocity changes of slightly better than two parts in 10^4 .

Attenuations were determined by comparing successive echo heights as displayed on an oscilloscope with the output of an adjustable RC generator also displayed on the same oscilloscope. Minimum accuracy of the dial calibration was ± 0.002 dB cm^{-1} below 2.4 dB cm^{-1} , and the reproducibility of a given setting was always better than ± 0.02 dB cm^{-1} .

Temperature control was accomplished by pumping on liquid oxygen which is a more suitable cryogenic fluid than liquid nitrogen for the particular temperature range. All measurements were taken with the specimen just immersed in boiling liquid oxygen. The specimen temperature was determined by a platinum resistance wire thermometer placed in contact with the specimen. Resistance was measured to within three milliohms which gives an accuracy of ± 5 millidegrees based upon published data.¹²

Figure 1 shows the attenuation of longitudinal waves of various frequencies propagating parallel to [110] in MnF_2 . The attenuation peak amplitude increases with frequency and to within the accuracy of the experiments does not shift in temperature. The peak occurs at $67.35 \pm 0.02^\circ\text{K}$, which

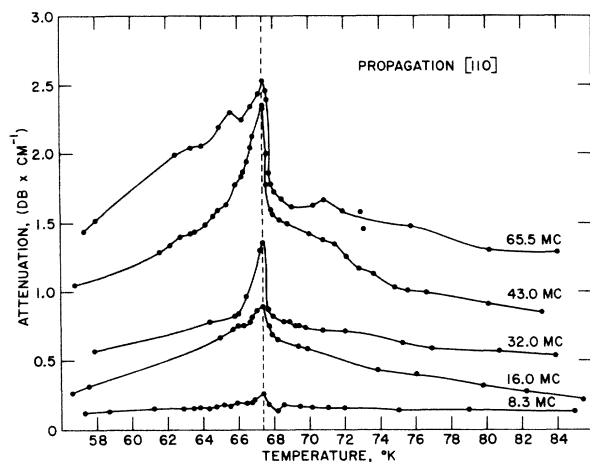


FIG. 1. Attenuation versus temperature for longitudinal waves propagating in the $[110]$ direction in MnF_2 . The maximum attenuation occurs at 67.35°K for all frequencies.

is slightly removed from the Néel temperature of 67.336°K determined by Heller and Benedek.¹³

Details of the attenuation peak for 25-Mc/sec longitudinal waves are shown in Fig. 2, where the attenuation is shown on an expanded temperature scale. The solid circles are data taken with zero applied magnetic field and complement the data of Fig. 1. The open circles and crosses are data taken in a 3.6-kG applied field oriented perpendicular and parallel to the c axis, respectively. It is apparent that the magnetic field does not affect the amplitude or position of the attenuation peak. However, the magnitude of attenuation immediately adjacent to the Néel temperature was decreased slightly with applied magnetic field, causing an apparent narrowing of the attenuation peak. The attenuation peak for the longitudinal waves occurred at 67.35°K for all frequencies measured. The maximum value observed was over 2.6 dB cm^{-1} . Although the peak is approximately of the same magnitude as that observed for MnO and CoO , no large accompanying change in modulus is observed. (The modulus changes observed in MnO and CoO at lower frequencies were approximately 50%.)

For shear waves, the attenuation increases with frequency, but no peak was observed near the Néel temperature for waves polarized either parallel or perpendicular to the c axis. The effect of a 5.5-kG external magnetic field was to change the general attenuation level slightly (0.1 dB cm^{-1}) with field orientation.

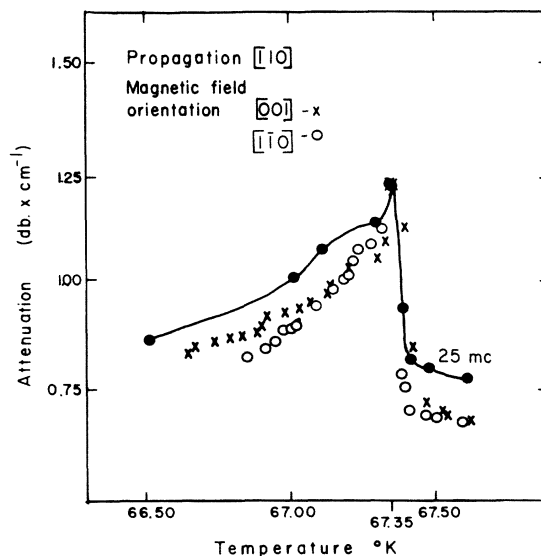


FIG. 2. Attenuation as a function of temperature for 25-Mc/sec longitudinal waves propagating parallel to $[110]$ in MnF_2 . The solid circles are for zero extended field. Open circles and crosses are for 3.6 kG applied magnetic field.

No change in acoustic wave velocity was observed for each of the three polarizations when the sample temperature was lowered through the Néel temperature. Any velocity change is estimated to be less than one part in 5000, and a change in modulus is apparent only through the thermal expansion. For MnF_2 , the change in length along the a axis first decreases and then increases¹¹ with decreasing temperature, being zero at 80°K . Thus the change in modulus through the Néel temperature occurs almost entirely through the density change resulting from thermal expansion parallel to the c axis. For a temperature change from 70°K to 60°K , this change amounts to about three parts in 10^4 .

The transition in MnF_2 at 67°K is thought to be an order-disorder phase change between the two orientations of spins. The thermodynamics of second-order phase changes¹⁴ predicts that the thermal expansion coefficient, specific heat, and isothermal compressibility of an isotropic solid should all become large at the transition temperature. From the measurements taken here, it is apparent that any large changes in the compressibility must come about through changes in elastic constants other than those measured. Experiments to determine this effect, principally through changes in C_{33} , and the effects of larger magnetic fields are being continued.

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XENON IMPURITY STATES IN SOLID ARGON*

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Recent measurements on solid rare gas films¹ have revealed unexpected absorption bands tentatively interpreted^{1,2} in terms of Wannier³ excitons. The present note reports the observation of similar "trapped excitons" in an Ar film containing small amounts of Xe. Thus the previous interpretation is supported, and, moreover, the first experimental determination of the effective mass of a conduction electron in a solid rare gas is achieved.

Films of argon containing various small percentages of Xe were prepared by deposition from a gas mixture onto a cold substrate, and the optical absorption spectra of the films were obtained using essentially the equipment previously described.¹ The temperatures involved ranged from 8 to 10°K and concentrations in the original gaseous mixtures ranged from 1 to 0.1 mole percent. Some enrichment in the Xe concentration is expected because of its lower vapor pressure, and we estimate that this enrichment is of the order of a factor of 3. Figure 1 shows the optical density of a film Ar:~0.003Xe between photon energies of 8 and 12 eV, a region which is featureless and generally transparent in pure argon films.¹ On the basis of estimated oscillator strengths in pure Xe,⁴ it can be deduced that the film thickness in this experiment was ~5μ.

It is seen that a rich variety of absorption lines appear, in contrast to the simple pair of resonance lines in atomic Xe at 8.43 eV and 9.57 eV.

These resonance lines belong to transitions of the type $5p^6(^1S_0) - 5p^56s(^3P_1, ^1P_1)$. The absorption bands A_1 and B_1 are assumed to correspond to these atomic transitions, with a splitting in the solid of 1.31 eV due to a (somewhat enhanced)

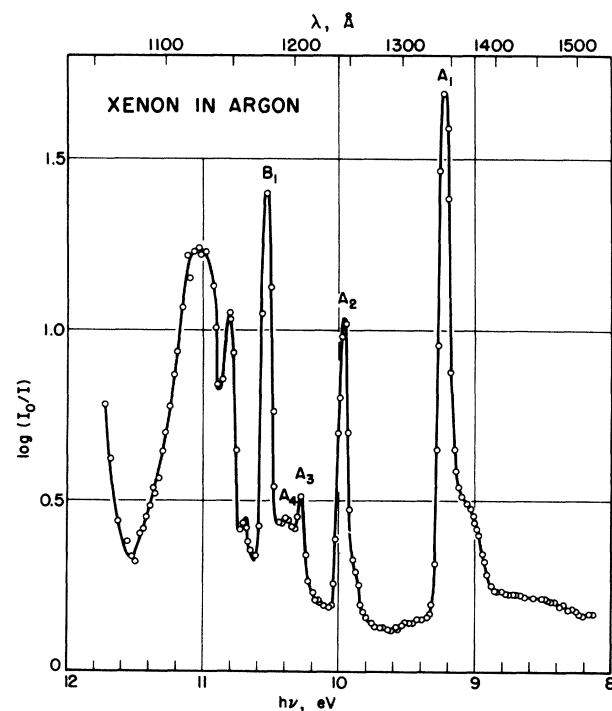


FIG. 1. Absorption spectrum of a film of Ar containing ~0.3 mole % Xe at 10°K.